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Author(s):
Stauffer, Fritz; Stoll, Sebastian; Kipfer, Rolf; Kinzelbach, Wolfgang

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Analytical model for environmental tracer transport in well catchments

Fritz Stauffer,[1] Sebastian Stoll,[1] Rolf Kipfer,[2,3] and Wolfgang Kinzelbach[1]

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[1] In this paper, we present analytical solutions and discuss them for simplified groundwater systems with decaying environmental tracers such as ^{3}H, including the formation of a decay product such as tritiogenic ^{3}He. The developed solutions are applicable for shallow, unconfined groundwater systems, which can be conceptually described by a steady-state, two-dimensional, semiconfined groundwater flow model with constant thickness, recharge rate, and porosity. The prerequisite for the applicability of our solutions is that the pumping wells and observation wells at which tracer information is available are fully screened over the entire aquifer thickness. The sampling by pumping from such wells produces the complete mixing of water of different age and origin. The application of our solution to the Baltenswil (Zurich, Switzerland) groundwater field site shows that the simplified model is able to catch essential dynamics of the transient concentration development of ^{3}H and ^{3}He.

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1. Introduction

[2] Environmental tracer data are increasingly used in groundwater hydrology. They are often used to support a direct interpretation of specific flow conditions. For example, tracer concentrations may indicate recent recharge, or they may even allow a quantitative estimate of the age of groundwater or its residence time. Furthermore, such data can yield streamline information, or information on the ratio of fluxes including recharge rates. If water fluxes in aquifer systems are known, tracer data can be translated into effective porosity values.

[3] Groundwater dating on the basis of simultaneous measurement of ^{3}H and tritiogenic ^{3}He was suggested by *Tolstikhin and Kamensky* [1969] and applied in the field of groundwater hydrology by a large number of investigators such as *Poreda et al.* [1988], *Schlosser et al.* [1988], *Schlosser et al.* [1989], *Solomon et al.* [1992], *Szabo et al.* [1996], *Cook and Herczeg* [1999], *Holocher et al.* [2001], and others. The method is based on the formula for the residence time as a function of the relative concentration of the two tracers in the saturated part of the aquifer:

$$t_{\mathrm{residence}} = \frac{1}{\lambda}\ln\left(\frac{c_{^3\mathrm{He}}}{c_{^3\mathrm{H}}}+1\right). \quad (1)$$

[4] The coefficient λ is the decay constant ($\lambda = \ln(2)/T_{1/2}$, where $T_{1/2}$ is the half-life of tritium). The formula is based on a moving small water parcel concept with negligible mixing and continuous accumulation of tritiogenic ^{3}He. Point sampling along a flow line allows estimates of the travel time. The ^{3}H-^{3}He age was used by, e.g., *Sheets et al.* [1998] to evaluate groundwater flow models using the particle tracking method. Among others, *Weissmann et al.* [2002] discussed the effect of dispersion on groundwater age, which is caused by water mixing.

[5] Analytical solutions of the transport equation and lumped-parameter or box models are still the prominent, and sometimes the only, tools for the interpretation of environmental tracer data in terms of residence times [*Vogel*, 1967; *Vogel*, 1970; *Maloszewski and Zuber*, 1982, 1983; *Leibundgut et al.*, 2009; *Cook and Böhlke*, 1999]. These models are usually based on the assumptions that the groundwater system is at steady state, sufficiently homogeneous, and is subject to well-defined recharge and extraction conditions. Lumped-parameter models are characterized by specific transit time distributions. New developments to determine the residence time distribution use time series of environmental tracer concentrations and interpret them by nonparametric deconvolution methods [*Cirpka et al.*, 2007].

[6] A further important application of environmental tracer data is to define and select conceptual groundwater models and to improve numerical groundwater flow and transport models, or to determine solute transport parameters [*Wei et al.*, 1990; *Reilly et al.*, 1994; *Zoellmann et al.*, 2001; *Mattle et al.*, 2001; *Castro and Goblet*, 2003; *Zuber et al.*, 2005; *Troldborg et al.*, 2007]. The integration of environmental tracers into groundwater models can reduce the range of possible alternative interpretations, which are all consistent with all observations.

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[1] Institute of Environmental Engineering, ETH Zurich, Zurich, Switzerland.

[2] Department of Water Resources and Drinking Water, EAWAG, Duebendorf, Switzerland.

[3] Institute of Mineralogy and Petrography, ETH Zurich, Zurich, Switzerland.

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[7] However, one has to be aware that there exists a series of limitations for the use of environmental tracers in groundwater modeling. For example, the input function is not always well known, or the age window of a particular tracer is too restrictive. Most environmental tracer data do not provide direct information on Darcy fluxes. Dissolved gas tracers yield information, which is different from that of solute markers of the water molecule (e.g., ^{3}H). Estimated porosity values are notoriously subject to uncertainty. Moreover, the effective porosity may not be constant because of dual porosity effects. Above all, one has to remember that hydraulic heads usually represent a momentary situation, while tracer data integrate flow conditions over longer time periods. The residence time of tracers in the unsaturated zone can sometimes be larger (even much larger) than the residence time in the saturated zone of the aquifer [e.g., *Onnis*, 2007]. Long-lasting unsaturated transport can increase the uncertainty, since information on unsaturated flow conditions is usually very vague. Nevertheless, the range of possible results obtained through flow modeling can sometimes be restricted considerably by using environmental tracer information. This restriction depends on the sensitivity of a particular parameter such as porosity with respect to environmental tracer concentrations. There are examples in which transmissivity is not, or is only barely, sensitive with respect to environmental tracer transport [e.g., *Onnis*, 2007]. An important problem is posed by the mixing of water of different age and origin, even in relatively small groundwater volumes.

[8] In this paper, analytical solutions are derived and discussed for simplified shallow groundwater systems with radioactively coupled environmental tracers, e.g., decaying ^{3}H, including the production and accumulation of the decay product, e.g., tritiogenic ^{3}He. The simplified system consists of a two-dimensional, semiconfined aquifer model with constant thickness, recharge rate, and porosity. The idea behind choosing a two-dimensional depth-averaged approach is the fact that pumping wells or observation wells in shallow aquifers are often fully screened over the complete aquifer thickness. Whenever sampling is undertaken using the pump in the case of abstraction wells, or sampling pumps in the case of the observation well, the complete screening implies the mixing of water of different age and origin in wells, where the samples for age analysis are taken. The approach is conceived as a simplified base-model case to assess the behavior of environmental tracers in well and spring catchments.

2. Mathematical Development

[9] Groundwater flow is modeled for two-dimensional, steady-state, and semiconfined conditions assuming constant aquifer thickness H and constant recharge rate N. The flow equation in this case is

$$\nabla \cdot (T \nabla h) + N = 0, \tag{2}$$

where T is the aquifer transmissivity. No assumptions are made with respect to spatial variability of T. The flow domain is arbitrary and consists of the catchment of one pumping well or spring. Tracer transport is modeled for both steady-state and transient conditions with constant and transient input functions. A consequence of the vertical integration in the two-dimensional model formulation with steady-state flow field and recharge is the fact that concentrations at a given location always represent average values with contributions from many different stream tubes, as indicated in Figure 1. Such an averaging process has implications for the tracer concentrations measured in fully screened pumping wells and fully screened observation wells.

2.1. Solution for Exponentially Decreasing Input Function of Decaying Species

[10] The two-dimensional transient transport equation for a decaying species with transient input (recharge) concentration $c_{\text{in}}(t)$ at the groundwater table is

$$\frac{\partial c}{\partial t} = \nabla \cdot (\mathbf{D} \nabla c) - \nabla \cdot (\mathbf{u}\, c) + \frac{N\, c_{in}}{\phi H} - \lambda c, \tag{3}$$

where $\mathbf{D}$ is the dispersion tensor (including molecular diffusion and macrodispersion effects), $\mathbf{u}(\mathbf{x})$ is the velocity vector, $c(\mathbf{x})$ is the concentration of the decaying species, ϕ is the effective aquifer porosity, and $c_{\text{in}}(t)$ is the transient input (recharge) concentration of the decaying species. No assumptions are made with respect to spatial variability of $\mathbf{D}$ and $\mathbf{u}$. Effective porosity ϕ is taken as constant. Using the flow equation (2), the transport equation (3) can be reformulated as follows:

$$\frac{\partial c}{\partial t} = \nabla \cdot (\mathbf{D} \nabla c) - \mathbf{u} \nabla c + \frac{N}{\phi H} \cdot (c_{\text{in}} - c) - \lambda c. \tag{4}$$

[11] The boundary conditions are

$$(u_{\text{n}} \cdot c)_{\text{x\_boundary}} = 0 \tag{5}$$

or

$$(\partial c / \partial n)_{\text{x\_boundary}} = 0, \tag{6}$$

where u_{n} is the velocity component normal to the boundary and n is the direction normal to the boundary at location

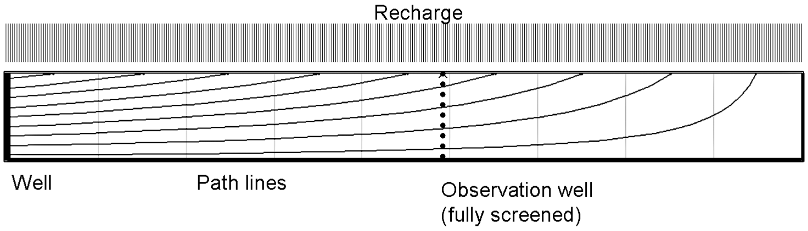

Figure 1. Schematic cross section of a shallow aquifer with recharge and path lines.

$\mathbf{x}_{\text{\_boundary}}$. The transient input function $c_{\text{in}}(t)$ of the decaying species is assumed to be an exponentially decreasing function of time:

$$c_{\text{in}}(\mathbf{x}, t) = c_{\text{in},0} \exp(-\gamma t), \tag{7}$$

where $c_{\text{in},0}$ is the initial input (recharge) concentration at time $t = 0$ and γ is a constant decay coefficient of the tracer input function. The initial condition of the aquifer concentration $c(\mathbf{x}, t = 0)$ is

$$c(\mathbf{x},\ t = 0) = c_0\,, \tag{8}$$

where c_0 is the initial aquifer concentration of the decaying species. The solution is

$$\begin{aligned} c(\mathbf{x}, t) &= \frac{c_{\text{in}}(t)}{\left[\frac{(\lambda-\gamma)\,\phi H}{N} + 1\right]} = \frac{c_{\text{in},0} \exp(-\gamma t)}{\left[\frac{(\lambda-\gamma)\,\phi H}{N} + 1\right]} \\ &= \frac{c_{\text{in},0} \exp(-\gamma t)}{[(\lambda - \gamma)\, t_{\text{residence}} + 1]} \end{aligned} \tag{9}$$

and

$$c_0 = \frac{c_{\text{in},0}}{[(\lambda - \gamma)\, t_{\text{residence}} + 1]}. \tag{10}$$

[12] The solution is independent of location $\mathbf{x}$. This means that the concentration of the decaying species is homogeneously distributed over the whole solution domain, depending on the aquifer parameters ϕ, H, and N. The term $(\phi H \lambda)/N$ is related to the mean residence time of the water in the saturated zone of the well/spring catchment:

$$t_{\text{residence}} = \frac{\phi H}{N}. \tag{11}$$

[13] From equation (9), it follows that the residence time can be evaluated for given input and aquifer concentrations at any location $\mathbf{x}$ provided that $\gamma \neq \lambda$. For $\gamma = \lambda$, the solution is

$$c(\mathbf{x}, t) = c_{\text{in}}(t) = c_{\text{in},0} \exp(-\gamma t). \tag{12}$$

[14] This means that the solution in this case is independent of aquifer parameters and residence time.

[15] For $\gamma = 0$, the solution for constant tracer input $c_{\text{in}}(t)$ is obtained from equation (9). Accordingly, the tracer concentrations are constant throughout the domain. *Vogel* [1970] found an identical expression to equation (9) for the mean concentration of spring water with constant tracer input concentration (for $\gamma = 0$), homogeneous hydraulic conductivity, and constant aquifer thickness and recharge rate. Note that our solution does not assume constant transmissivity. The same solution also corresponds to the analytical solution of *Maloszewski and Zuber* [1982] for the lumped-parameter model of decaying species with exponential weighting function and constant input concentration. Our model therefore allows for specifying flow and transport conditions of a physical model, for which this particular lumped-parameter model holds.

[16] A typical application of the solution is the ^{3}H evolution with exponentially decreasing or constant input concentrations, provided that the discussed assumptions are *at least approximately* fulfilled.

2.2. Solution for the Decay Product of a Decaying Species With Exponentially Decreasing Input Function

[17] The two-dimensional transient transport equation for the decay product (concentration $c(\mathbf{x}, t)$) of a decaying species of concentration $c_{\text{dec\_sp}}$ with exponentially decreasing input (recharge) concentration at the groundwater table is

$$\begin{aligned} \frac{\partial c}{\partial t} = {} & \nabla \cdot (\mathbf{D}\,\nabla c) - \mathbf{u}\,\nabla c + \frac{N}{\phi H} \cdot (c_{\text{in}} - c) \\ & + \lambda\, c_{0,\text{dec\_sp}} \exp(-\gamma t)\,, \end{aligned} \tag{13}$$

where $c_{0,\text{dec\_sp}}$ is the initial aquifer concentration of the decaying species. The concentration c_{in} is the input (recharge) concentration of the decay product, which is assumed to obey $c_{\text{in}} = 0$ here. Still, the same boundary conditions (equations (5) and (6)) hold. The solution is

$$c(\mathbf{x}, t) = \frac{c_{0,\text{dec\_sp}} \exp(-\gamma t)}{\left[\frac{N}{\phi H \lambda} - \frac{\gamma}{\lambda}\right]} = \frac{c_{0,\text{dec\_sp}} \exp(-\gamma t)}{\left[\frac{1}{t_{\text{residence}}\,\lambda} - \frac{\gamma}{\lambda}\right]}, \tag{14}$$

which again is independent of location $\mathbf{x}$. The condition for a meaningful solution is

$$\frac{1}{t_{\text{residence}}\,\lambda} - \frac{\gamma}{\lambda} > 0 \text{ or } t_{\text{residence}} < \frac{1}{\gamma}. \tag{15}$$

[18] In terms of the input (recharge) concentrations $c_{\text{in,dec\_sp}}$(t) of the decaying species, the solution is

$$\begin{aligned} c(\mathbf{x}, t) &= \frac{c_{\text{in\_0,dec\_sp}} \exp(-\gamma t)}{\left[1 - \frac{2\gamma}{\lambda} + \frac{N}{\phi H \lambda} - \frac{\phi H \gamma}{N} + \frac{\phi H \gamma^2}{N\lambda}\right]} \\ &= \frac{c_{\text{in\_0,dec\_sp}} \exp(-\gamma t)}{\left[1 - \frac{2\gamma}{\lambda} + \frac{1}{t_{\text{residence}}\,\lambda} - t_{\text{residence}}\gamma + t_{\text{residence}}\frac{\gamma^2}{\lambda}\right]}. \end{aligned} \tag{16}$$

[19] The ratio of the concentrations of the decaying species and the decay product is

$$\frac{c_{\text{dec\_sp}}(\mathbf{x}, t)}{c(\mathbf{x}, t)} = \frac{N}{\phi H \lambda} - \frac{\gamma}{\lambda} = \frac{1}{t_{\text{residence}}\,\lambda} - \frac{\gamma}{\lambda}. \tag{17}$$

[20] Therefore, the residence time can be expressed by

$$t_{\text{residence}} = \frac{1}{\lambda \cdot \left[\frac{c_{\text{dec\_sp}}(\mathbf{x},t)}{c(\mathbf{x},t)} + \frac{\gamma}{\lambda}\right]}. \tag{18}$$

[21] Note that the residence time is again independent of location $\mathbf{x}$.

[22] For $\gamma = 0$, the solution for constant tracer input $c_{\text{in}}(t)$ is obtained from equations (14) or (16). Accordingly, the tracer concentrations are constant throughout the domain. This holds true also for the residence time. Equation (16) with $\gamma = 0$ corresponds to the solution of *Maloszewski and Zuber* [1983] for the corresponding lumped-parameter model of the tritiogenic ^{3}He production in groundwater.

[23] A typical application of the solution is the tritiogenic ^{3}He system with exponentially decaying ^{3}H input function, provided that all assumptions are *sufficiently* fulfilled and $t_{\mathrm{res}} < 1/\gamma$, which is about 18 years.

2.3. Transient Solution for Linearly Increasing Input Function of Decaying Species

[24] The transient transport equation is given by equation (4). The transient input function of decaying species (recharge) concentration is linearly increasing with time according to

$$c_{\mathrm{in}}(\mathbf{x}, t) = c_{\mathrm{in},0} + \alpha t. \qquad (19)$$

[25] In this case, the solution for constant aquifer thickness, porosity, and recharge rate and using equation (11) is

$$c(\mathbf{x}, t) = \frac{c_{\mathrm{in}}(t) - \frac{\alpha t_{\mathrm{residence}}}{(\lambda t_{\mathrm{residence}} + 1)}}{(\lambda t_{\mathrm{residence}} + 1)} = \frac{c_{\mathrm{in}}\left(t - \frac{t_{\mathrm{residence}}}{(\lambda t_{\mathrm{residence}} + 1)}\right)}{(\lambda t_{\mathrm{residence}} + 1)}. \qquad (20)$$

[26] This allows for estimating the residence time for given input (recharge) concentration and given aquifer concentration at any location $\mathbf{x}$. A possible application of the model would be the ^{85}Kr system with approximately linearly increasing input concentrations, provided that the assumptions are *sufficiently* fulfilled.

3. Application to the Baltenswil Site

[27] The application of our solutions to the Baltenswil site is given as an illustration and confrontation with field data. The well catchment of the drinking water pumping well Baltenswil (Zurich, Switzerland) is situated within the highly conductive sandy gravel Aatal aquifer in the upper Glatt valley. The aquifer is unconfined and is laterally delimited and covered by moraines. The aquifer bottom consists of loamy lake sediments, which are considered as impermeable. It is naturally drained by a series of springs, and the domain contains no rivers or creeks. Groundwater is abstracted in several wells (Figure 2). The pumping wells and all observation wells are fully screened over the complete aquifer thickness. The Baltenswil well catchment covers an area of approximately 1.5 km^2 (based on steady-state flow considerations). Land use in the well catchment includes agriculture and forestry to about equal parts and a small part consists of settlements. The available measurements in the pumping wells and observation wells show that the mean saturated sandy gravel aquifer thickness H is 5.9 m and does not vary much (PW Baltenswil: 5.5 m; Kb94–1: 5.5 m; Kb94–2: 6.5 m; Kb14: 6.1 m). However, the measurement locations do not include the lateral moraine part of the catchment, where it can be assumed that the aquifer thickness is much smaller. The mean recharge rate in the catchment is estimated as 1.3 mm/d for the period 1999–2006, based on precipitation, temperature, and radiation measurements at a nearby meteorological station (Zurich-Kloten, at a distance of 6 km) and evaluation of the actual evapotranspiration rate. The estimated range because of land use variability is between 1.2 and 1.6 mm/d. Porosity ϕ is estimated at about 0.2 for this type of formation [*Jussel et al.*, 1994]. In the following, we assume the parameters H and ϕ to be constant over the sandy gravel aquifer area of the catchment.

[28] The tritium input concentration in precipitation is shown in Figure 3 for the stations Konstanz (Germany, data from IAEA, available at http://www-naweb.iaea.org/napc/ih/IHS_resources_isohis.html, 2009), Vaduz (Principality of Liechtenstein), and Basel (Switzerland, the latter two data sets from *BAG* [2009]). The closest station is Konstanz at a distance of 50 km. The tritium decay curve in Figure 3 suggests that, essentially, the input curve can be approximated by the natural tritium decay curve for the period after 1998. Therefore, the approximation $\gamma \cong \lambda$ holds and equations (12) and (16) are applicable, provided that the residence times in the unsaturated and the saturated zones

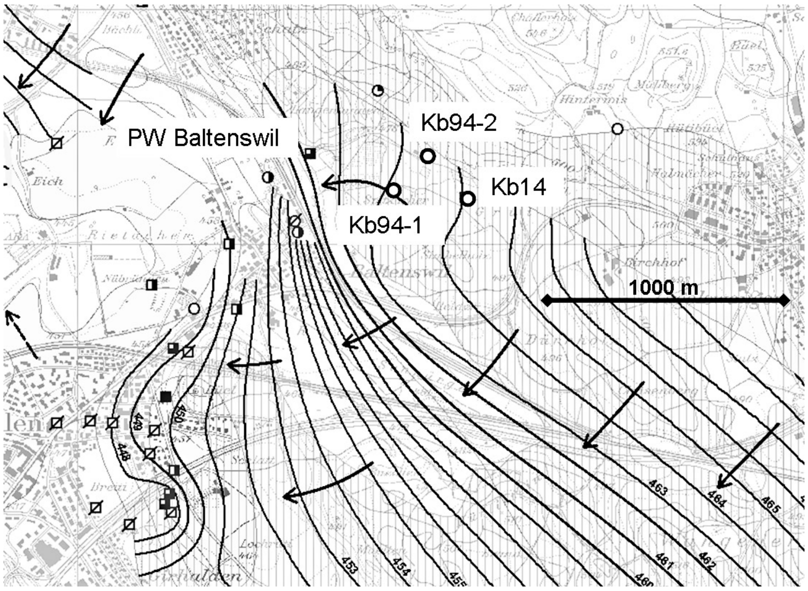

Figure 2. Groundwater map with head contours of the Baltenswil site with location of pumping wells (squares) and observation wells (circles). (Groundwater map [*AWEL*, 2010]).

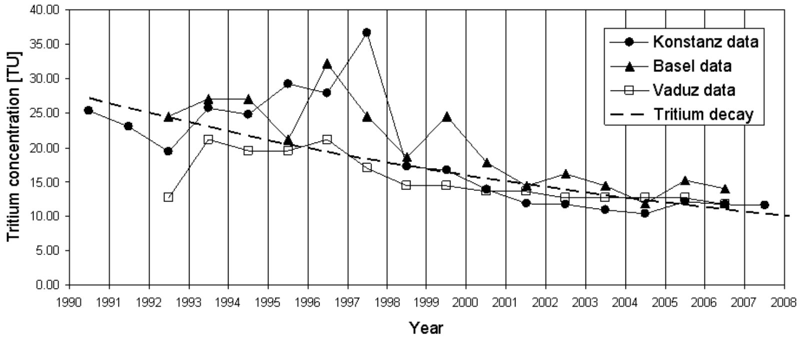

Figure 3. Measured tritium concentration in precipitation, together with tritium decay function.

are small enough. The travel time in the unsaturated zone was estimated to be of the order four years [*Onnis*, 2007].

[29] The concentrations of ^{3}H and tritiogenic ^{3}He were measured in the pumping well Baltenswil and in the observation wells in irregular time intervals between 2000 and 2006. The ^{3}H and tritiogenic ^{3}He concentration data for all stations and all samples are shown in Figure 4. A table with the tracer concentrations can be found in the work of *Onnis* [2007]. Figure 4 also includes fitted ^{3}H and ^{3}He decay curves according to our developed solutions in equations (12) and (16). Both curves fit all data relatively well and concentrations are essentially within the experimental error bandwidth over the investigation period.

[30] The residence time during the period 2000 and 2006 was calculated according to equation (18). The results are shown in Figure 5. Within the experimental error, the residence time remains approximately constant over the 6 years, with some fluctuations for all measurement locations. Note that, for the observation wells, the residence time is a representative value for the term $(\phi H)/N$. The average residence time over all locations is 1.54 years and for the Baltenswil well alone it is 1.55 years. The residence time in the aquifer is therefore small. Using equation (1) to evaluate the residence time according to an averaged moving parcel concept yields similar values (1.6 years in the mean). By taking the mean decay function for tritium with $\gamma = \lambda$, and evaluating equation (17) using the two decay curves in Figure 4, the term $N/(\phi H \lambda)$ assumes the value of 12.6, which relates recharge rate to effective porosity and aquifer thickness. Therefore, one of the three parameters can be determined if the other two are given. With an effective porosity of 0.2 and a mean aquifer thickness of 5.9 m (for the sandy gravel part of the catchment), the recharge rate would be 2.3 mm/d. This is higher than the estimated mean recharge rate from precipitation data and actual evapotranspiration calculation because the effective aquifer area without considering the lateral moraine part is smaller than the total mean recharge area, which includes the moraine. Since, in the moraine part, the saturated thickness is unknown but is expected to be smaller, the mean overall saturated thickness over the complete catchment would be reduced, which consequently would lead to a smaller recharge rate.

4. Discussion and Conclusions

[31] In this paper, analytical solutions are presented and discussed for simplified shallow groundwater systems with radioactively coupled environmental tracers, e.g., decaying ^{3}H, including the production and accumulation of a decay product, e.g., tritiogenic ^{3}He. The case with linearly increasing input function would be an approximate model for the evaluation of tracers such as ^{85}Kr. Note that the solutions can also be used for conservative nondecaying tracers such as SF$_6$. The developed solutions are applicable for shallow unconfined groundwater systems, which can reasonably be approximated by a steady-state, two-dimensional, semiconfined aquifer flow and transport model with constant thickness, recharge rate, and porosity. The model further assumes that the pumping wells and observation wells are fully screened over the complete aquifer thickness. This is fulfilled for many shallow groundwater systems. Whenever sampling is undertaken using the pump in the case of abstraction wells, or sampling pumps in the case

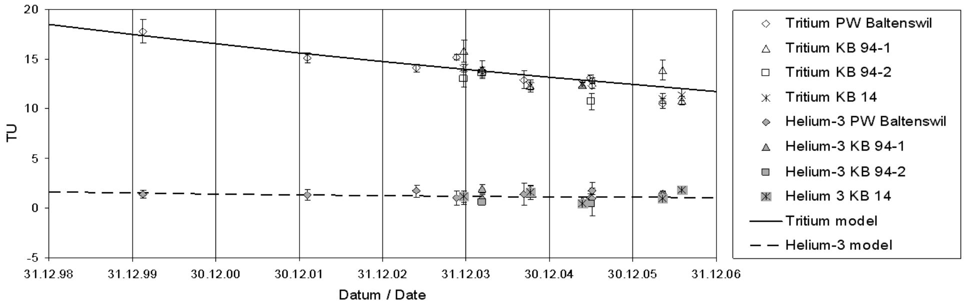

Figure 4. Tritium and helium-3 concentrations with error bars, together with modeled concentrations using equations (12) and (16).

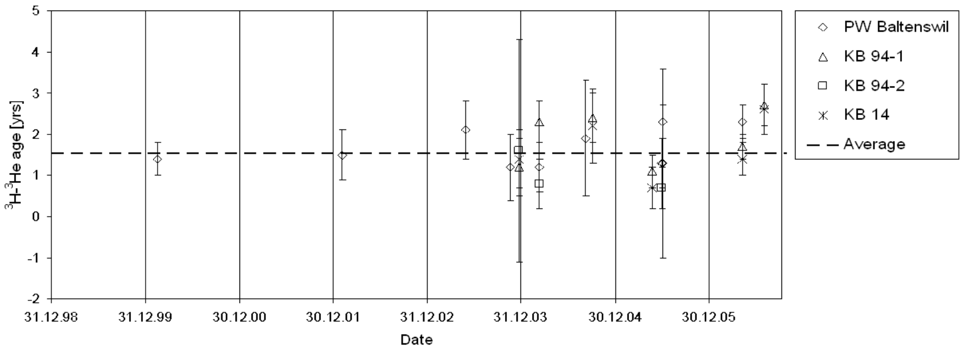

Figure 5. Tritium-helium-3 age according to equation (18) with error bars.

of observation wells, the complete screening implies the mixing of water of different age and origin in wells where the samples for age analysis are taken. Under these conditions, our solutions predict that the transient decaying and accumulating environmental tracer concentrations are homogeneously distributed in the model domain, provided that the assumptions are satisfied. Our solutions provide relations between aquifer thickness, porosity, and recharge rate for given tracer concentrations. Moreover, for the ^{3}H-^{3}He system, the residence time can be determined for given tracer concentrations.

[32] In the case of the Baltenswil catchment, the developed transient solution with exponentially decaying input function is able to successfully model the temporal evolution of the ^{3}H and ^{3}He concentrations. Since the transient ^{3}H input function approximately equals the ^{3}H decay curve with $\gamma = \lambda$, the ^{3}H data alone yield no information about the residence time or other any aquifer parameters. This conclusion is in line with the observations by *Onnis* [2007], who found virtually no sensitivity of the tritium transport to transmissivity and only little sensitivity to porosity in numerical transient flow and transport modeling of the same aquifer. The conclusion can also be of interest for other applications.

[33] Deviations from our analytical solutions are expected if the simplifying assumptions are not fulfilled in a specific application. For instance, the assumptions of constant aquifer thickness and constant recharge rate are very often not valid. Also, the steady-state flow assumption is frequently violated. However, in some cases, these parameters might be replaced by average values, as it seems to be the case for the Baltenswil catchment. If this concept holds, acceptable results can be expected for a wide range of applications. The presented relationship between the parameters N, H, and ϕ can be used to estimate one of the three parameters if the other two are given. Usually, one would use it to estimate the mean recharge rate of shallow unconfined porous aquifers. Moreover, the model can be very useful for testing prevailing concepts of the aquifer system. The presence of a homogeneous apparent age distribution within the aquifer for ideal conditions is a further interesting property. The solutions are supposed to break down whenever strong lateral inflows of water with different composition of environmental tracer concentrations occur or strongly inhomogeneous recharge concentrations are present. In such cases, a numerical model would be indispensable. Moreover, deviations from the solutions are also expected for strongly heterogeneous hydraulic conductivity values, where the classical advection-dispersion equation is no longer valid. Nevertheless, the presented approach represents simplified base-model cases, which are suited to assess the behavior of environmental tracers in well and spring catchments of shallow unconfined groundwater systems.

[34] **Acknowledgments.** The sampling and measurement of environmental tracers for the Baltenswil catchment was performed within the Swiss National Science Foundation project 200,020-1074489/1 entitled "Models and tracer methods for groundwater modeling." The formulation of the analytical solutions and the evaluation of the data were partially supported by the EU seventh Framework Programme GENESIS project 226,536.

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W. Kinzelbach, F. Stauffer, and S. Stoll, Institute of Environmental Engineering, ETH Zurich, Wolfgang-Pauli-Str. 15, CH-8093 Zurich, Switzerland. (stauffer@ifu.baug.ethz.ch)

R. Kipfer, Department of Water Resources and Drinking Water, EAWAG, CH-8600 Duebendorf, Switzerland.